

Generation, Disposition, and Current Inventory of Radionuclides in the INTEC Tank Farm

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June 2005

**Idaho
Cleanup
Project**

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U.S. Department of Energy by CH2M ♦ WG Idaho, LLC

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Idaho Falls, Idaho 83415

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ABSTRACT

This report provides an inventory of the major, long-lived, radionuclide activity in the INTEC Tank Farm. It includes the cumulative amount of activity that has been sent to, removed from, and is currently stored in the 300,000-gallon tanks. It also provides a Tank Farm activity removal percentage, based upon the activity remaining in the tanks and the activity originally sent to the tanks. This data is needed to fulfill the requirements of the 2005 Defense Authorization Bill (S. 2400) Section 3116 Defense Site Acceleration Completion that stipulates radioactive waste determination criteria based partly upon the removal of radionuclides from the Tank Farm.

The Tank Farm radionuclide inventory is based upon the most recent waste samples and includes the activity in both the solid and liquid phases. Radionuclides not measured in waste samples are estimated using ORIGEN2-based models. This report incorporates recent revisions of the ORIGEN2-based models used to estimate radionuclide activity for which sample data do not exist. The revisions to the estimate models change the activity estimates from that of other recent radionuclide inventory reports.

SUMMARY

This report provides an inventory of the long-lived, radionuclide activity in the INTEC Tank Farm. It includes the cumulative activity of radionuclides that have been sent to, removed from, and is currently stored in the 300,000-gallon tanks. It also provides a Tank Farm activity removal percentage, based upon the activity remaining in the tanks and the activity originally sent to the tanks.

The radioactivity inventory is decayed to January 1, 2012, the approximate future closure date of the Tank Farm facility. The use of decayed activity data reduces the percentage of the radioactivity removed from the tanks because it does not include the activity that was removed from the tanks, calcined, and decayed in the calcined storage facilities. The overall removal percentage for all of the radionuclides combined is 98.5%. Removal and treatment of the remaining waste will increase the final removal percentage to over 99.9%. The removal percentage of most of the individual radionuclides is in the upper 90% range, similar to that of the overall removal percentage. However, a few radionuclides have lower removal percentages. These include short-lived species (Pu-236) for which the calculated removal percentage is lower than the actual because the activity removed many years ago is not counted due to its decay. It also includes some species (U-238) whose source (pilot plant studies) and waste storage history (as sodium-bearing waste) differed from that of most radionuclides. Consequently, it was not removed with the bulk of the waste and activity that was calcined.

The radioactivity inventory data is needed to fulfill the requirements of the 2005 Defense Authorization Bill (S. 2400) Section 3116 Defense Site Acceleration Completion that stipulates radioactive waste determination criteria based partly upon the removal of radionuclides from the Tank Farm. Similar data have been reported in several previous reports for purposes such as the Environmental Impact Statement, Waste Incidental to Reprocessing determination, and future sodium-bearing waste treatment studies. The activity in such reports has varied due to differences in the waste inventory over time, assumptions in the mass and composition of undissolved solids, and the radionuclide estimate models. This report discusses the reasons for the differences in the radionuclide inventory differences between this and other recent reports.

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Generation, Disposition, and Current Inventory of Radionuclides in the INTEC Tank Farm

1 INTRODUCTION

This report provides a radionuclide inventory for the waste originally sent to, currently stored in, and removed from the Idaho Nuclear Technology and Engineering Center (INTEC) Tank Farm. The data in this report will help fulfill the requirements of the 2005 Defense Authorization Bill (S. 2400) Section 3116 Defense Site Acceleration Completion that stipulates radioactive waste determination criteria based partly upon the removal of radionuclides from the Tank Farm.

Over the past few years, several reports have documented the INTEC Tank Farm waste inventory. Although the reported radioactivity varied, the reports typically did not include data comparison tables or explanations for the differences. The reported activity varied due to varying assumptions, sample data availability, changes to the models used to estimate radionuclide activity, and changes in waste inventory due to waste calcination etc. This report details the reasons for the radioactivity variation among several recent reports. It also provides a current inventory of the radioactivity in the Tank Farm based upon the most recent Tank Farm data, including the volume of liquid waste, mass of undissolved solids, waste sample analyses, and revised ORIGEN2-based estimates for radionuclides for which sample data do not exist. Much of this data, especially the ORIGEN2-based estimates differ from those of previous reports, resulting in changes in the estimated Tank Farm radionuclide inventory.

The radionuclide inventory in this report was compiled in a Microsoft Excel data base. The data base can be easily updated to incorporate new or revised data for liquid waste volumes, mass of undissolved solids, waste sample analyses, ORIGEN2-based radionuclide estimates, or radionuclide decay dates to provide an updated radionuclide inventory in the future. A copy of the data base is on the CD included with this report.

2 INTEC TANK FARM RADIONUCLIDE INVENTORY BACKGROUND

The INTEC Tank Farm has provided interim storage of radioactive wastes for about 50 years. During that time many of the tanks were filled with waste, emptied when the waste was calcined, and then refilled. As a result, the bulk of the activity sent to the Tank Farm has either been removed and calcined or decayed in storage. The waste remaining in the Tank Farm is called sodium-bearing waste and contains only a small fraction of the activity originally sent to the Tank Farm. Several recent reports have documented the radioactivity remaining in the Tank Farm. However, due to varying assumptions and operation of the Calcliner, the reported activity has varied among the reports. This section provides historical information on the source and disposition of Tank Farm waste and an explanation of the variation in the reported Tank Farm radionuclide inventory among recent reports.

2.1 INTEC Tank Farm and Waste History

Irradiated nuclear fuel has been reprocessed at the Idaho National Laboratory (INL) since 1953 using facilities located at the Idaho Nuclear Technology and Engineering Center (INTEC), formerly known as the Idaho Chemical Processing Plant (ICPP). Historically, INTEC received spent nuclear fuel (SNF) from a variety of reactors throughout the world and stored it underwater in pools or in dry storage facilities. Some of the SNF was chemically reprocessed to recover highly enriched uranium and other nuclear-related products for the Department of Energy (DOE) and its predecessor organizations. SNF reprocessing and its related support activities produced liquid waste that was stored in the Tank Farm. Most of the liquid waste has been removed from the Tank Farm and solidified using a process called calcination. The calcination process evaporated the water from the liquid waste and converted the dissolved constituents (including radionuclides) into dry granular solids. The solids produced by this process, called calcine, are stored in calcined solids storage facilities (CSSFs), which consist of stainless steel storage bins contained in concrete vaults.

Fuel reprocessing produced two general types of liquid waste. One type is known today by its regulatory term of high-level waste (HLW). The other type is a transuranic (TRU) waste that is currently referred to as sodium-bearing waste (SBW). Generally, these wastes were stored separately in the Tank Farm due to differences in chemical composition, heat generation and cooling requirements, and subsequent processing (calcination) requirements.

Typically, SNF reprocessing began by dissolving the fuel (cladding, fission products, and uranium) in acid. Highly enriched uranium was recovered from the acidic dissolver product in a liquid extraction system. The extraction process produced an aqueous, acidic waste stream, called first-cycle raffinate (known today as HLW), which contained the dissolved fuel cladding and the bulk of the radioactive fission and activation products. A variety of SNF types were reprocessed at INTEC. The chemical composition of the resulting waste depended on the type of fuel and chemicals used during reprocessing. Aluminum (Al) and zirconium (Zr)-clad fuels were the most frequently reprocessed fuels and consequently produced the greatest volumes of waste. Smaller quantities of stainless steel and graphite matrix fuel were also processed, and each produced a chemically unique waste stream.

SBW came from a variety of sources. These included the second and third-cycle uranium extraction processes (where the uranium recovered from the first-cycle extraction process was purified), equipment decontamination, laboratories, off-gas condensate, ion exchange systems, etc. The name “sodium-bearing” waste came from the relatively high sodium ion concentration (1-2 molar) in the waste. The high sodium concentration was the result of activities that made extensive use of sodium-based chemicals such as sodium hydroxide and sodium carbonate. SBW has had a variety of names. Since most of the

SBW was concentrated by the Process Equipment Waste (PEW) Evaporator prior to storage in the Tank Farm, it has also been called PEW Evaporator concentrate or “bottoms”. The radionuclide content of SBW was typically much lower (over an order of magnitude) than HLW.

From 1953 to 1992, SNF was routinely reprocessed and the resulting liquid waste was stored in the Tank Farm. The INTEC Tank Farm provided interim storage for the waste. Typically, a tank was filled with waste, sampled, and then emptied by transferring the waste to a calcining facility, where the liquid waste was converted to a solid granular form. From 1963 to 1981, liquid waste was treated in the Waste Calcining Facility (WCF). From 1982 to 2000, liquid waste was treated in the New Waste Calcining Facility (NWCF), which replaced the original WCF.

In April 1992, DOE announced it would no longer reprocess spent fuel at INTEC and called for a shutdown of the INTEC reprocessing facilities. Since that time, no more HLW has been generated from SNF reprocessing. The NWCF treated the last of the HLW that had been stored in the Tank Farm in February 1998. Calcination of SBW continued through May 2000 when it ceased in compliance with a Consent Order¹.

For all practical purposes, only SBW remains in the Tank Farm. Although the Calciners treated all types of Tank Farm waste, they processed HLW faster and more efficiently than SBW. Consequently, all of the HLW was calcined before the shutdown of the Calciner in May 2000, leaving only SBW in the Tank Farm. When a 300,000-gallon tank that had contained HLW was “emptied”, 4000 to 12,000 gallons of waste remained in the bottom of the tank due to the configuration of the tank and waste transfer piping (the tanks have flat bottoms and do not have a bottom drain line). The waste remaining in an empty tank was called the tank “heel”. The HLW heels that remained in the emptied tanks no longer exist as a distinct volume or type of waste. Some of the HLW tanks were refilled with large volumes (over 250,000 gallons) of SBW. The small quantity of HLW heel can no longer be distinguished from the large volume of SBW. In other tanks, the HLW heel was flushed from the tank. The flushing process added water and/or SBW to a tank and transferred the heel/flush solution mixture from the tank. Several consecutive flushes effectively removed the HLW heel from the tank. The flush solution was typically concentrated by evaporation and then transferred into a tank containing SBW. Loos (2004) provides a detailed history of the Tank Farm, its waste, waste sources, waste disposition, etc. Loos (2004) estimates that approximately one percent of the waste remaining in the Tank Farm is derived from HLW, primarily from the HLW heels left in emptied tanks.

For the purpose of this report, the Tank Farm radionuclide inventory is the SBW contained in three 300,000-gallon tanks (WM-187 through WM-189) and the small amount of contaminated water in WM-190. Tanks WM-187, -188, and -189 are full, or nearly full, and contain about 830,000 gallons² of concentrated SBW. WM-190 has never been used to store highly contaminated or concentrated waste. It has been maintained as an “empty” spare to receive waste from other tanks should a leak develop. WM-190 contains about 500 gallons of water (rainfall and snowmelt) that were transferred from the tank vault into the tank in the 1960s to keep the empty tank from floating in the water that accumulated in the vault. The water is contaminated as a result of waste leaking through the valves isolating the tank from the main waste transfer lines, or from occasional cycling of the isolation valves (to verify their position), which allowed waste to drain from the transfer lines into the tank.

¹ Third Modification of the Notice of Noncompliance Consent Order, dated April 20, 1999.

² The waste volume in the tanks is measured by two independent monitoring systems. The systems have a zero offset and thus provide slightly differing values for the waste volume. This report uses the waste volume from the Digiquartz volume measurement system.

Other tanks originally associated with the Tank Farm have been emptied and cleaned, or are being used by other processes. Four 18,000-gallon tanks located in the CPP-604 tank vaults (WL-101 and WM-100, -101, and -102) remain in use as part of the PEW Evaporator system. The four 30,000-gallon tanks (WM-103 through -106) and seven of the 300,000-gallon tanks (WM-180 through -186) are empty and no longer in service. The empty tanks have been cleaned as part of a RCRA tank closure effort. Separate reports are being written to document the small amount of residue (rinse water and solids) and the radioactivity remaining in those tanks after cleaning activities are complete. The small amount of residue and the activity in those tanks is not included in the waste inventory of this report.

2.2 Recent Tank Farm Radionuclide Inventory Report Comparisons

Several recent reports (Millet and Staiger 2000, Swenson 2003, and Barnes 2004,) documented the radionuclide inventory of the SBW stored in the INTEC Tank Farm. In general, each of those reports used waste sample (analytical) data as the basis for the radionuclide inventory when such data were available. They used ORIGEN2-based models to estimate the activity of radionuclides for which sample data did not exist. However, the Tank Farm radionuclide activity differed, often significantly, among the reports. The reported Tank Farm radionuclide inventory varied because the purpose and assumptions, available sample data, and Tank Farm waste inventory differed for each report.

Millet and Staiger (2000) provided a conservatively high radionuclide inventory for the Tank Farm for use in the Environmental Impact Statement or EIS (DOE 2002) to provide a bounding inventory for accident scenario calculations. For example, Millet and Staiger (2000) used the same radionuclide source term for the undissolved solids in every tank, regardless of the type of waste or solids in the tank. The solids source term came from the historical solids sample having the (conservatively) highest radionuclide activity. That source term was used even for solids in tanks for which lower-activity, historical sample data existed (Swenson 1992). When estimating the activity of radionuclides for which sample data did not exist, Millet and Staiger (2000) conservatively used the highest ORIGEN2-based activity estimate from among five different radionuclide estimate models, instead of using the model for SBW. For example, they used the Pu-238 activity from the Zr waste model and the Pu-239 activity from the electrolytic waste model because the Zr and electrolytic waste models had higher estimated activities for those radionuclides than the SBW model. As a result, the EIS basis report contained a conservatively high, bounding estimate of the radioactivity in the Tank Farm.

Swenson (2003) documented the radionuclide source term used for the waste incidental to reprocessing (WIR) determination. The WIR and EIS reports were based upon the same volume of Tank Farm waste (measured in July 1999). However, the Tank Farm activity in the WIR report was much lower than in the EIS report. The WIR report used less conservative assumptions than the EIS to estimate the radionuclide activity in the Tank Farm. The WIR report applied one of three radionuclide source terms to the solids in the tanks, instead of a single source term, based upon the waste storage history of the tanks. It applied the high-activity source term used by the EIS to the solids in only three of the tanks, and it reduced the mass of solids in two of those three tanks from the amount used in the EIS. The WIR report applied one of two lower-activity source terms to the solids in the other tanks. The WIR report consistently used the ORIGEN2-based model for SBW to estimate those radionuclides for which sample data did not exist, instead of the EIS method of using the maximum value from among the five different waste models for each radionuclide. As a result of reducing the conservative assumptions, the Tank Farm radionuclide inventory in the WIR report was significantly less than the EIS report, about 60% less for Cs-137 for example.

Barnes (2004) provided a Tank Farm radionuclide source term for use as a SBW feed composition in a future waste treatment process. The Tank Farm radionuclide inventory in the SBW feed composition report is generally less than the inventory in the WIR or EIS reports because it was based upon the Tank Farm waste inventory that existed after the Calciner finished operating in May 2000. The Calciner operation during 2000 removed approximately 25% of the Tank Farm radioactivity inventory (Swenson 2003). The SBW feed composition report included the operation of the Calciner during 2000, some of the waste concentration (evaporation) and consolidation that occurred after the Calciner operation, and waste sample data obtained after July 1999 that were not available for the EIS and WIR reports. The SBW feed composition report increased the estimate of the total mass of solids in the tanks (based on operational experience) from that of the WIR and EIS estimates. Although the increase in the estimate of the mass of solids increased the radionuclide inventory in the solids portion of the waste, the activity reduction due to the operation of the Calciner in 2000 resulted in a net decrease in the total Tank Farm activity, compared to the EIS and WIR reports.

This report provides an updated Tank Farm radionuclide inventory. The inventory differs from that in the SBW feed composition report for several reasons. The SBW feed composition report included estimates for some “future” operations, such as the concentration of WM-180 waste, that have been completed and are included in this report. This report includes recent sample data that did not exist at the time the SBW feed composition report was written. This report makes minor changes (less than 5%) in the estimate of the mass of solids in the Tank Farm. The most significant change in this report is the use of data from a recent revision (Wenzel 2005) of the ORIGEN2-based estimates of the radionuclides in various types of INTEC wastes. The new ORIGEN2-based estimates do not significantly change the activity of most of the major fission products. However, the new estimates change the activities of most of the activation products and transuranic (TRU) elements. For example, the new estimates change the activity of the major TRU components (Pu-238, Am-241, etc.) by a factor of about two in the SBW. The new estimates include more significant changes to some of the minor TRU components (such as Cm-248), up to an order of magnitude in some cases.

In addition to providing the current Tank Farm radionuclide inventory, this report also documents the total radioactivity of the major, long-lived radionuclides that were added to the Tank Farm since it was placed in service and the activity that has been removed from the tanks (calcined or shipped from INTEC). These data were included in the WIR report (Swenson 2003), but were not in the SBW feed composition report. This information is necessary for the section 3116 waste determination process.

3 TANK FARM RADIONUCLIDE ACTIVITY BALANCE

Historically, the Tank Farm provided interim storage for a variety of wastes. The waste inventory in a given tank was not static over time. Most tanks were filled with waste, emptied when the waste was calcined, and refilled with more waste. Some tanks went through this filling/emptying cycle several times. Radionuclides were removed from the Tank Farm by three mechanisms. Most of the waste (about eight million gallons) and radioactivity were removed from the Tank Farm and calcined. The radionuclides in that waste are now in storage in the CSSFs. In the 1950s and early 1960s, relatively small amounts of waste (totaling about 15,000 gallons) were removed from the tanks and shipped off site. In addition, much of the radioactivity sent to the Tank Farm, especially that of the short-lived species, has decayed in storage.

Figure 1 is a simplified block diagram showing the activity of selected radionuclides that entered the Tank Farm and the disposition of that activity (shipped offsite, calcined, or remaining in the Tank Farm). The data in Figure 1 are based upon the Tank Farm waste inventory in April 2005, with the radionuclides decayed to January 1, 2012 (approximate closure date of the Tank Farm). In keeping with the method of the WIR report (Swenson 2003), Figure 1 does not include radionuclides that decayed in storage. Using decayed activities conservatively understates the activity of most of the radionuclides that were sent to and removed from the Tank Farm. This conservatism is important to the waste determination which depends on the removal of radioactivity. For example, Figure 1 shows about 9.5 million Ci of Cs-137 were sent to the Tank Farm (activity decayed to 2012). Actually, about 23 million Ci of Cs-137 were originally sent to the Tank Farm (Tyson 2002), but over half of it will have decayed by 2012, resulting in the reported value of 9.5 million Ci. Most of that activity was removed from the tanks, calcined, and decayed within the CSSFs. The decay of short-lived species such as Ru-106, Zr-95, and Ce-144 is even more pronounced than Cs-137. The activity of Ce-144 was originally higher than Cs-137 in some of the waste. However, Ce-144 has a half life of less than 1 year and its activity has decayed to virtually nothing in both the calcine and the remaining Tank Farm waste. Short-lived radionuclides (excluding short-lived daughters of long-lived radionuclides) are not included in this report. Including the original activity of the short-lived radionuclides would increase (by several times) the activity sent to the Tank Farm, the activity calcined or decayed in storage, and the percentage of the radionuclides removed from the Tank Farm. Reporting decayed activities and excluding short-lived species conservatively understates the total activity originally sent to and removed from the Tank Farm.

This report provides the radionuclide activity remaining in the four 300,000-gallon tanks that remain in service (WM-187 through WM-190). Residual activity in the tanks that have been emptied and cleaned is not included in either the total activity generated or the activity remaining in the Tank Farm. The residual activity in the cleaned tanks is being reported elsewhere and that data is not repeated in this report. Samples of the residual liquid and solids indicate the Cs-137 activity in a cleaned 300,000-gallon tank is a few hundred curies. Compared to the total Cs-137 activity generated (about 9.5 million Ci) and the Cs-137 currently remaining in the tanks (about 150 thousand Ci), the residual activity in cleaned tanks is insignificant to the mass balance in Figure 1 and is within the error of the activity estimates of this report.

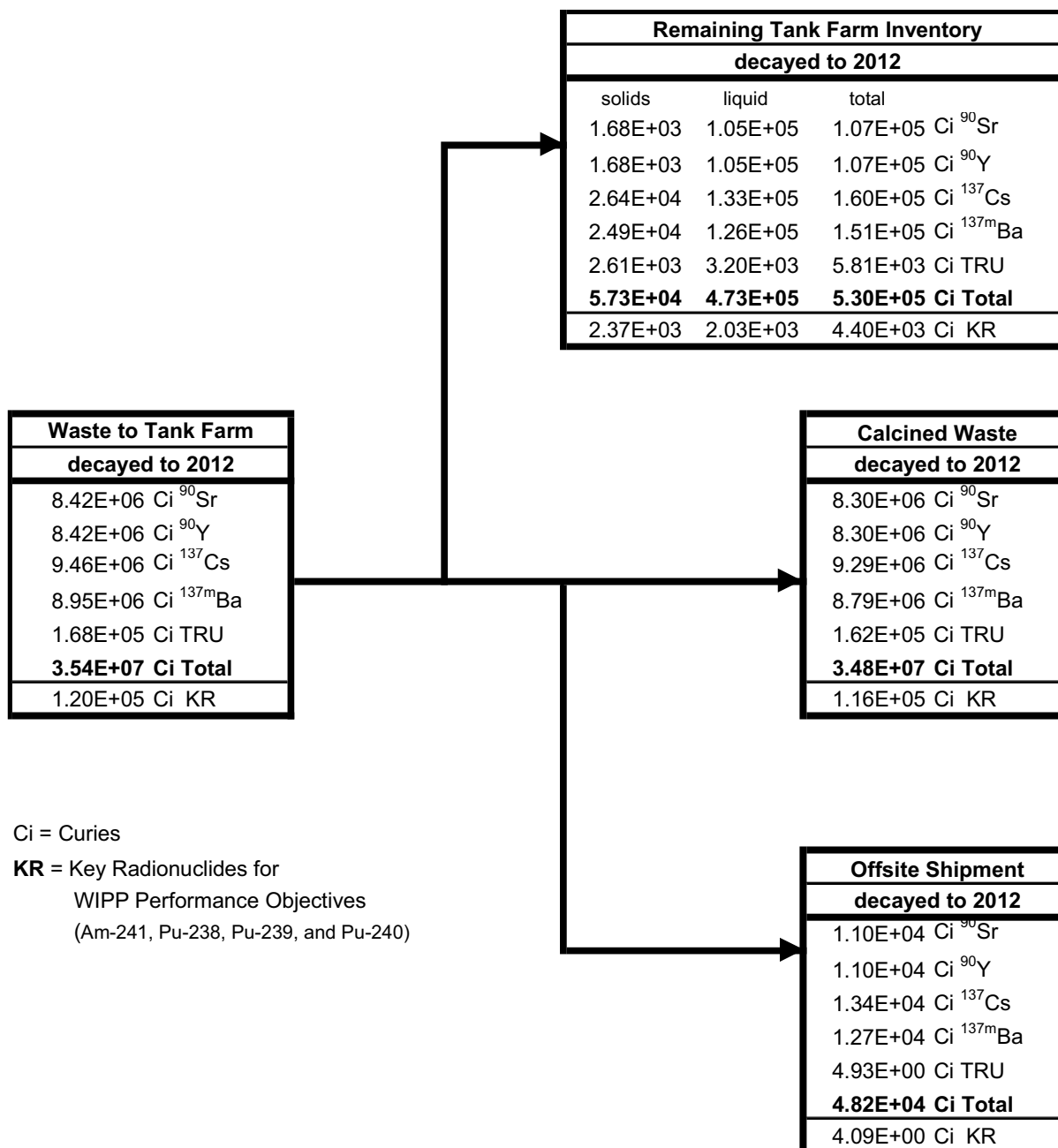


Figure 1. Block diagram showing the activity sent to, removed from, and remaining in the Tank Farm.

Figure 1 does not include all of the radioactive elements in the Tank Farm. It includes those species responsible for the bulk of the activity (⁹⁰Sr/⁹⁰Y and ¹³⁷Cs/^{137m}Ba) and the elements deemed most significant to long-term waste disposal and management (TRU elements). The radionuclides in Figure 1 account for 99% of the radioactivity in the tanks. Figure 1 does not include tritium (H-3), ¹³⁴Cs, and other radionuclides that account for about 1% of the total Tank Farm activity. Many of those elements are included in more detailed tables elsewhere in this report. Figure 1 includes a group of radionuclides termed “key radionuclides” (KR) that includes Pu-238, Pu-239, Pu-240, and Am-241. These radionuclides were singled out for consistency with a similar mass-balance block diagram in the WIR report (Swenson 2003). The KR activity is included in both the “TRU” and “total” radionuclide

categories of Figure 1. Their activity is given separately due to their significance to the Waste Isolation Pilot Plant (WIPP) performance objectives.

3.1 Radionuclide Activity Data Sources

The radioactivity in the Tank Farm, CSSFs, and off-site shipments shown in Table 1 came from a combination of waste sample (analytical) data and ORIGEN2-based estimates. In general, waste sample data was used when available. Analytical data for the major fission products (such as Cs-137 and Sr-90) and TRU elements (such as Pu-238 and Pu-239) were available for much of the current and historical waste inventory. For less significant radionuclides and those present in trace amounts (such as Cm-248) analytical data were not available and ORIGEN2-based estimates of the activity were made.

Samples of Tank Farm wastes have been taken and analyzed for both chemical and radionuclide content since the Tank Farm was placed in service in the 1950s. Hundreds of Tank Farm waste samples have been analyzed over the years. However, until recently there was no systematic effort made to collect and retain the analytical data for more than a few years or to generate a comprehensive database of the sample data. Beginning in the 1990s a diligent effort was made to retrieve and compile historical Tank Farm analytical data for use in estimating the composition of the calcine in the CSSFs. The effort to compile detailed data on the composition of Tank Farm waste was limited by the ability to locate old sample data, and the scope of the historical sample analyses. Data were gathered from numerous sources including published reports and letters, microfilmed historical data files, historical analytical laboratory files, and employee personal files. This effort generated a large database of information on the chemical and radiochemical content and the volumes of historical Tank Farm wastes that were calcined (Staiger and Swenson 2005).

Typically, historical sample analyses focused on those species needed for corrosion control, heat generation calculations, shielding calculations, dose calculations, and other parameters needed to manage the waste. Trace components that had no effect on waste management were not included in the sample analyses. For example, data on radionuclides such as Sr-90, Cs-137, and significant, short-lived gamma emitters were available on virtually every waste sample because they were important for calculations involving shielding, heat generation, hypothetical accident dose calculations, etc. On the other hand, analytical data for trace components such as curium (Cm) generally do not exist because such elements were present in such small quantities that they were not significant to the management of the waste, or could not be detected by available laboratory methods.

Some elements for which historical analytical data do not exist may be significant to long-term waste management. Therefore, computer models were developed to estimate nuclides in various types of Tank Farm wastes. The models used ORIGEN2 (Croff 1980) to generate data on fission and activation products produced in a reactor. Because INTEC reprocessed different types of nuclear fuels that produced differing wastes, ORIGEN2-based models were developed for each of the major types of fuel reprocessed at INTEC. Models were developed for Zr, Al, and stainless steel-clad fuels. A model was also developed for coprocessing waste, which was produced from simultaneously reprocessing Zr and Al-clad fuels and blending the resulting dissolver products.

An ORIGEN2-based computer model was also developed for SBW by treating it as a “pseudo” fuel type. A primary source of SBW was the decontamination of HLW processing equipment. Decontamination solution generally had the same relative ratios of fission products as HLW, but in significantly smaller quantities. The SBW model was developed by blending the waste compositions predicted by a combination of the Al, Zr, and stainless-steel-clad fuel models until the relative fission product activity of

the resulting combination matched SBW sample data. SBW also contained significant quantities of second and third-cycle waste from the uranium purification process. Second and third-cycle wastes contained relatively large concentrations of TRU elements due to the chemistry of the SNF uranium extraction and purification process. The models for all waste types were adjusted by multiplying the activity of the TRU components predicted by ORIGEN2 by an adjustment factor so that the final value predicted by the computer model matched sample data of the various types of wastes.

The activities of radionuclides for which analytical data do not exist were estimated using one of the five waste models (Al, Zr, stainless steel, coprocessing, or SBW). This was done by using Cs-137 as a predictor for the relative activity of other radionuclides. Trace or unanalyzed nuclides in a given waste were calculated by multiplying the Cs-137 activity in the waste of interest by the nuclide-to-Cs-137 ratio from the selected waste model. Cs-137 was used as the predictor because it is easily and accurately measured in Tank Farm wastes. Historical Cs-137 data exist on virtually all of the hundreds of historical Tank Farm waste samples, making it possible to calculate nuclide quantities for Tank Farm wastes. Historically, the composition of Tank Farm waste was maintained to prevent the separation or partitioning of radionuclides (such as solids precipitation). There were generally no INTEC processes downstream of the Tank Farm that selectively concentrated any particular nuclide, so the relative radionuclide ratios remained constant for a given waste stream. Exceptions to this include H-3, C-14, and I-129, which formed volatile compounds at elevated temperatures and were emitted as gases when subjected to the Calcliner or PEW Evaporator processes. As a result, the Cs-137 activity is a good predictor of the activity of most radionuclides.

This report uses recently revised ORIGEN2-based radionuclide estimates (Wenzel 2005) for the various types of INTEC wastes that differ from the activity estimates used in recent Tank Farm inventory reports (EIS, WIR, and SBW feed composition). The ORIGEN2-based models were revised to incorporate recommended changes (Swenson 2004a and Swenson 2004b) to improve the accuracy of the estimates. The revision corrected a few errors noted in previous models, made the various waste models more consistent in their assumptions and methodology, and incorporated a revised SBW “adjustment basis” source term (the Tank Farm sample data used to adjust the ORIGEN2 estimates). The model revisions generally made minor changes to the activity estimates of the fission products. The most significant changes were to the estimates of some of the activation and TRU components. The estimated activity of the major TRU components (Pu-238 and Pu-239) in the SBW model changed by a factor of up to 3. However, the estimate of the activity of a few of the minor TRU components (Cm-248 and Cf-251) and activation products (Ni-63) changed by an order of magnitude or more.

The revised radionuclide estimate model significantly increased (4 to 5 orders of magnitude) the activity of carbon 14 (C-14) in Zr waste (and consequently coprocessing and SBW) from that of the previous model. The estimated C-14 activity increased because of the large amount of contaminant nitrogen assumed to be in the Zr alloy fuel cladding in the Zr waste model. A review (Swenson 2005) of available data showed this resulted in an unrealistically high C-14 activity in the Tank Farm wastes. Therefore, the Tank Farm radionuclide inventory in this report is based upon the C-14 estimates of the previous ORIGEN2-based waste models, and the revised C-14 estimates were not used.

3.2 Radioactivity Generated and Sent to the Tank Farm

The “Waste to Tank Farm” box on Figure 1 contains the activities of selected radionuclides sent to the Tank Farm between 1953 and April 2005, decayed to January 1, 2012. Very little additional activity is expected to be added to the Tank Farm in the coming years, so the total activity generated is not expected to change significantly. The only expected significant change to Figure 1 is the future treatment of the

SBW that will remove the bulk of the activity currently in the Tank Farm. The radionuclide activity in the “Waste to Tank Farm” block is the sum of the activities in the Figure 1 blocks for the activity in the CSSFs, shipped off site, and remaining in the Tank Farm.

Figure 1 shows only selected radionuclides, corresponding to those in a similar block diagram in the WIR report. It includes the activities of Cs/Ba-137m, Sr/Y-90, and the TRU components, which account for 99% of the total activity in the waste and include those species considered important to future facility closure activities. Short-lived nuclides such as Ru-106, Ce-144, Zr-95, etc. are not included in Figure 1.

Figure 1 includes only the activity of waste that was stored in the 300,000-gallon tanks. It does not include radionuclides that were stored in the CPP-604 waste tanks (WM-100, -101, and -102) or the 30,000-gallon tanks (WM-103 through -106) and were never stored in the 300,000-gallon tanks. Those tanks were used to segregate some wastes (such as stainless steel and ROVER wastes) that were transferred directly to the Calciners for treatment and were never stored in the 300,000-gallon tanks. Because such activity is not included in either the waste generated or waste calcined, the calcined waste activity in this report differs slightly from that of other reports (Staiger and Swenson 2005).

3.3 Radionuclides Shipped Offsite

The data in the “Offsite Shipment” box on Figure 1 came from a compilation of “CPP Production Weekly Reports” documenting the operating history of INTEC from 1955 to 1960 (Swenson 2003). In December 1955, a sample station was installed in tank WM-180. At the time, WM-180 was being filled with first-cycle raffinate from the dissolution of Al-clad fuel. From 1955 through 1960, waste was taken from WM-180, placed in 500-gallon transfer casks, and shipped to Oak Ridge National Laboratory (ORNL). ORNL removed radionuclides from the waste to build a national inventory of radionuclides. Table 1 lists the volumes of the waste shipments sent to ORNL and the CPP weekly report numbers from which the waste shipping information was obtained. The weekly reports usually specified the volume of waste shipped. However, some reports had less precise values, such as “part of a container”, which was interpreted as 250 gallons, or half the container volume.

Table 1. Waste volumes shipped offsite.

CPP Report Number	Volume (Gallons)
Rei-343-55A	200
Rei-15-56A	212
Rei-7-58A	480
Rei-54-58A	490
Rei-100-58A	485
Rei-192-58A	1,455
Rei-215-58A	970
Rei-227-58A	2,900
Rei-249-58A	1,455
Rei-273-58A	1,455
Rei-153-59A	2,500
Rei-185-59A	1,500
Ay-9-60A	1,250
Total	15,352

There are no sample analyses available for the wastes sent to ORNL. The radionuclide composition of a WM-180 sample taken in 1960 was used as the composition of all of the waste shipped to ORNL. Tank WM-180 filled slowly throughout the time the ORNL sample shipments were made. The waste entering the tank during that time came from the dissolution of Al-clad fuel and did not change significantly while the tank was filled. Therefore, the 1960 WM-180 waste composition should adequately represent the composition of the waste sent to ORNL.

Figure 1 and Table 1 do not include a few very small (less than 10 gallons) shipments of waste. The quantities of such shipments are within the potential error of the volume of the shipments shown in Table 1. Also, Figure 1 does not include 3.81 Ci (5.4 kg) of Np-237 that was recovered and sent to the Savannah River Site in 1972 because the Np-237 recovery occurred in the SNF reprocessing facility before the waste entered the Tank Farm.

Overall, the effect on the Tank Farm inventory made by off-site waste shipments was minor (0.14% of the total Cs-137 in Figure 1). Therefore, minor inconsistencies in the volume of waste shipped offsite or the waste composition entering WM-180 are inconsequential.

3.4 Radionuclides in Calcine

The “Calcined Waste” box in Figure 1 shows the bulk of the radionuclide activity that entered the Tank Farm was removed, calcined, and is stored in the CSSFs. INTEC had two calcining facilities. The Waste Calcining Facility (WCF) operated from 1963 through 1981. The New Waste Calcine Facility (NWCF) replaced the WCF and operated from 1982 through 2000. The calcination process sprayed liquid waste into a heated fluidized bed. The water in the waste evaporated leaving the dissolved waste constituents (fuel cladding, radioactive components, etc.) as small granular solids (primarily in the form of oxides such as Al_2O_3 , SrO , etc.) called calcine. The calcination process was developed because the solid calcine required less storage space and was safer to store (calcine is not corrosive to the storage vessel) than liquid waste.

The radionuclide activity in the “Calcined Waste” category is the product of the volume of waste calcined through May 2000 and its radionuclide activity, decayed to January 2012. Though simple in concept, this calculation is complex due to the number of times the waste tanks were filled and emptied with various types and compositions of wastes. The volumes and compositions of nearly 90 different wastes were compiled to generate the calcine radionuclide inventory. Additional information on the amounts and compositions of the calcine waste and the radionuclide inventory of the CSSFs can be found in Staiger and Swenson (2005).

As previously noted, the radionuclide inventory in this report does not include waste from the CPP-604 or 30,000-gallon waste tanks that was sent directly to the calcining facilities. Thus the calcine activity in Staiger and Swenson (2005) is slightly larger than the calcine activity in this report, because it includes more waste than this report. In addition, minor changes to the activity of Am-241 and Pu-244 were made to the inventory reported in Staiger and Swenson to provide more accurate activity estimates.

3.5 Radionuclides Remaining in the Tank Farm

The box labeled “Remaining Tank Farm Inventory” in Figure 1 includes radionuclides stored in the Tank Farm in both the liquid and solid phases. The Tank Farm radionuclide data are based on the volume of

waste measured in April 2005, estimates of the mass of solids in the tanks from the SBW feed composition report (Barnes 2004), and recent liquid and solid sample analyses. The April 2005 radionuclide inventory shown on Figure 1 was decayed to January 2012.

Table 2 provides a more detailed listing of radionuclides and activity for the solids and liquids in each of the four 300,000-gallon tanks. It is the source of the Tank Farm inventory data given in Figure 1. The total radioactivity in the Tank Farm given in Table 2 is slightly higher (about 1%) than the total activity in Figure 1. This is because the “total” radionuclide activity in Figure 1 is sum of the species listed in the block diagram on Figure 1. Table 2 contains a more extensive radionuclide list than Figure 1 (including H-3, Cs-134, Co-60, Tc-99, etc.). The additional radionuclides in Table 2 increase its total inventory about 1% above that given in Figure 1. Table 2 does not have a complete list of all radionuclides in the waste, but it includes those deemed significant to future waste treatment and includes the bulk (over 99%) of the activity. The list of radionuclides on Table 2 is the same as that in the WIR report (Swenson 2003).

The liquid waste volume for each tank given in Table 2 is the volume of total waste (liquid plus solid) less the volume of the estimated solids. The liquid volume in Table 2 does not match the “liquid” volume given in most INTEC reports. This is because most reports present the total waste volume as the volume of liquid and ignore the volume of solids present in the tanks. The reported waste volume also varies slightly (up to a few thousand gallons) depending on which of two waste tank volume measurements one uses. The two measurement systems (Digiquartz and radiofrequency) have a zero offset and do not give the same volume. This report uses the Digiquartz system for its waste volume data.

3.5.1 Radionuclides in the Tank Farm Liquid

The radioactivity in the liquid phase in Figure 1 is the activity in the four 300,000-gallon waste tanks (WM-187 through WM-190) that are currently in service. Three of the tanks (WM-187, -188, and -189) store concentrated SBW. WM-190 has about 500 gallons of contaminated water. The activity in WM-190 is insignificant compared to the other three tanks (about 0.01% of the total Tank Farm activity). However, the WM-190 activity is included for consistency with the WIR report (Swenson 2003), which included the WM-190 water in its inventory.

The liquid radionuclide inventory is the product of the volume of liquid waste in the tanks and its radionuclide activity. The volume of liquid waste was calculated as the total volume of waste less the volume of solids estimated to exist in each tank. The total waste volume in the tanks is accurately and continuously monitored by installed instrumentation in the tanks. The waste activity is based upon sample data from each of the four waste tanks and ORIGEN2-based estimates for those radionuclides for which analytical data do not exist. The composition of most of the SBW has not changed significantly since the most recent waste samples were obtained and analyzed. Years of experience have shown the sampling and analysis program used for Tank Farm liquids provides an accurate composition of the liquid waste in the tanks. Therefore, there is a relatively high degree of confidence in the accuracy of the radionuclide inventory in the liquid portion of the waste.

The liquid waste in WM-187 was sampled in January 2005 (sample log numbers 050112-4 and 050112-5) when the tank contained 265,000 gallons of waste. Since the tank was sampled, the change in the waste volume has been insignificant (total waste volume in April 2005 was 267,000 gallons). The small amount of additional waste came from sources similar to the rest of the waste in WM-187 and did not significantly affect the composition of the waste. Therefore, the January 2005 liquid waste sample is representative of the waste in WM-187.

Table 2. Tank Farm radionuclide inventory of April 2005 waste decayed to January 1, 2012 (all activity in curies).

	WM-187			WM-188			WM-189			WM-190			Tank Totals			
Liquid Volume (gallons)	252,600			281,800			281,700			500			816,600			
Solids Mass (kg)		105,000	Total Liquid + Solids Activity		5,000	Total Liquid + Solids Activity		5,000	Total Liquid + Solids Activity			Total Liquid + Solids Activity		115,000		
Phase	Liquid	Solid		Liquid	Solid		Liquid	Solid		Liquid	Solid		Liquid	Solids	Total Activity	
Am-241	5.38E+01	2.44E+01	7.81E+01	6.88E+01	2.65E-01	6.90E+01	7.79E+01	2.65E-01	7.82E+01	3.53E-02		3.53E-02	Am-241	2.00E+02	2.49E+01	2.25E+02
Am-242	1.88E-02	1.73E-02	3.61E-02	4.54E-02	7.57E-04	4.62E-02	3.09E-02	7.57E-04	3.16E-02	1.03E-05		1.03E-05	Am-242	9.51E-02	1.88E-02	1.14E-01
Am-243	2.60E-02	2.38E-02	4.98E-02	6.26E-02	1.04E-03	6.36E-02	4.25E-02	1.04E-03	4.36E-02	1.43E-05		1.43E-05	Am-243	1.31E-01	2.59E-02	1.57E-01
Cm-242	1.56E-02	1.43E-02	3.00E-02	5.59E-02	6.27E-04	5.65E-02	4.88E-02	6.27E-04	4.94E-02	8.57E-06		8.57E-06	Cm-242	1.20E-01	1.56E-02	1.36E-01
Cm-243	3.40E-03	3.12E-03	6.51E-03	8.18E-03	1.36E-04	8.32E-03	5.56E-03	1.36E-04	5.70E-03	1.86E-06		1.86E-06	Cm-243	1.71E-02	3.39E-03	2.05E-02
Cm-244	1.86E-01	1.71E-01	3.56E-01	8.68E-01	6.00E-03	8.74E-01	7.69E-01	6.00E-03	7.75E-01	1.02E-04		1.02E-04	Cm-244	1.82E+00	1.83E-01	2.01E+00
Cm-245	4.44E-05	4.07E-05	8.51E-05	1.07E-04	1.78E-06	1.09E-04	7.27E-05	1.78E-06	7.44E-05	2.44E-08		2.44E-08	Cm-245	2.24E-04	4.43E-05	2.68E-04
Cm-246	2.91E-06	2.67E-06	5.58E-06	7.01E-06	1.17E-07	7.13E-06	4.76E-06	1.17E-07	4.88E-06	1.60E-09		1.60E-09	Cm-246	1.47E-05	2.90E-06	1.76E-05
Cm-247	3.22E-12	2.96E-12	6.18E-12	7.76E-12	1.29E-13	7.89E-12	5.28E-12	1.29E-13	5.40E-12	1.77E-15		1.77E-15	Cm-247	1.63E-11	3.22E-12	1.95E-11
Cm-248	3.40E-12	3.12E-12	6.53E-12	8.20E-12	1.37E-13	8.33E-12	5.57E-12	1.37E-13	5.71E-12	1.87E-15		1.87E-15	Cm-248	1.72E-11	3.40E-12	2.06E-11
Np-237	5.57E-01	5.12E-01	1.07E+00	5.50E-01	3.21E-03	5.53E-01	4.96E-01	3.21E-03	4.99E-01	3.06E-04		3.06E-04	Np-237	1.60E+00	5.18E-01	2.12E+00
Pu-236	1.72E-04	1.58E-04	3.31E-04	4.15E-04	6.92E-06	4.22E-04	2.82E-04	6.92E-06	2.89E-04	9.46E-08		9.46E-08	Pu-236	8.70E-04	1.72E-04	1.04E-03
Pu-238	5.47E+02	1.95E+03	2.50E+03	6.33E+02	1.14E+01	6.44E+02	3.82E+02	1.14E+01	3.94E+02	3.00E-01		3.00E-01	Pu-238	1.56E+03	1.98E+03	3.54E+03
Pu-239	5.87E+01	3.54E+02	4.13E+02	8.05E+01	1.68E+00	8.22E+01	4.64E+01	1.68E+00	4.81E+01	3.22E-02		3.22E-02	Pu-239	1.86E+02	3.57E+02	5.43E+02
Pu-240	1.60E+01	1.47E+01	3.07E+01	3.86E+01	6.43E-01	3.92E+01	2.62E+01	6.43E-01	2.69E+01	8.78E-03		8.78E-03	Pu-240	8.08E+01	1.60E+01	9.68E+01
Pu-241	2.31E+02	2.12E+02	4.43E+02	5.57E+02	9.28E+00	5.66E+02	3.78E+02	9.28E+00	3.88E+02	1.27E-01		1.27E-01	Pu-241	1.17E+03	2.31E+02	1.40E+03
Pu-242	1.17E-02	1.07E-02	2.24E-02	2.82E-02	4.70E-04	2.87E-02	1.92E-02	4.70E-04	1.96E-02	6.42E-06		6.42E-06	Pu-242	5.91E-02	1.17E-02	7.08E-02
Pu-244	7.77E-10	7.13E-10	1.49E-09	1.87E-09	3.12E-11	1.90E-09	1.27E-09	3.12E-11	1.30E-09	4.26E-13		4.26E-13	Pu-244	3.92E-09	7.75E-10	4.69E-09
Cs-137	2.64E+04	2.42E+04	5.07E+04	6.36E+04	1.06E+03	6.47E+04	4.32E+04	1.06E+03	4.43E+04	1.45E+01		1.45E+01	Cs-137	1.33E+05	2.64E+04	1.60E+05
Ba-137m	2.50E+04	2.29E+04	4.79E+04	6.02E+04	1.00E+03	6.12E+04	4.09E+04	1.00E+03	4.19E+04	1.37E+01		1.37E+01	Ba-137m	1.26E+05	2.49E+04	1.51E+05
Sr-90	2.35E+04	1.24E+03	2.47E+04	4.92E+04	2.21E+02	4.94E+04	3.25E+04	2.21E+02	3.27E+04	1.29E+01		1.29E+01	Sr-90	1.05E+05	1.68E+03	1.07E+05
Y-90	2.35E+04	1.24E+03	2.47E+04	4.92E+04	2.21E+02	4.94E+04	3.25E+04	2.21E+02	3.27E+04	1.29E+01		1.29E+01	Y-90	1.05E+05	1.68E+03	1.07E+05
Tc-99	9.22E+00	2.30E+01	3.22E+01	2.84E+01	1.11E+00	2.95E+01	1.51E+01	1.11E+00	1.62E+01	5.06E-03		5.06E-03	Tc-99	5.27E+01	2.52E+01	7.80E+01
I-129	1.55E-02	1.43E-02	2.98E-02	3.74E-02	6.24E-04	3.80E-02	2.54E-02	6.24E-04	2.61E-02	8.53E-06		8.53E-06	I-129	7.84E-02	1.55E-02	9.39E-02
C-14	7.72E-05	7.08E-05	1.48E-04	1.86E-04	3.10E-06	1.89E-04	1.26E-04	3.10E-06	1.29E-04	4.23E-08		4.23E-08	C-14	3.89E-04	7.70E-05	4.66E-04
Cf-249	1.90E-11	1.75E-11	3.65E-11	4.58E-11	7.64E-13	4.66E-11	3.12E-11	7.64E-13	3.19E-11	1.04E-14		1.04E-14	Cf-249	9.61E-11	1.90E-11	1.15E-10
Cf-250	9.85E-12	9.04E-12	1.89E-11	2.37E-11	3.95E-13	2.41E-11	1.61E-11	3.95E-13	1.65E-11	5.41E-15		5.41E-15	Cf-250	4.97E-11	9.83E-12	5.95E-11
Cf-251	3.02E-13	2.77E-13	5.79E-13	7.27E-13	1.21E-14	7.39E-13	4.94E-13	1.21E-14	5.06E-13	1.66E-16		1.66E-16	Cf-251	1.52E-12	3.01E-13	1.82E-12
H-3	8.50E+00	1.90E-02	8.51E+00	1.11E+01	1.16E-03	1.11E+01	5.91E+00	1.16E-03	5.91E+00	4.66E-03		4.66E-03	H-3	2.55E+01	2.13E-02	2.55E+01
Co-60	1.57E+00	9.97E-01	2.56E+00	1.62E+01	1.17E-01	1.63E+01	1.63E+01	1.17E-01	1.64E+01	5.37E-03		5.37E-03	Co-60	3.41E+01	1.23E+00	3.53E+01
Ni-63	3.39E+01	3.11E+01	6.50E+01	7.11E+01	1.78E+00	7.29E+01	5.55E+01	1.78E+00	5.73E+01	1.86E-02		1.86E-02	Ni-63	1.60E+02	3.47E+01	1.95E+02
Se-79	2.83E-01	2.60E-01	5.44E-01	6.83E-01	1.14E-02	6.94E-01	4.64E-01	1.14E-02	4.75E-01	1.56E-04		1.56E-04	Se-79	1.43E+00	2.83E-01	1.71E+00
Sn-126	2.67E-01	2.45E-01	5.12E-01	6.43E-01	1.07E-02	6.54E-01	4.37E-01	1.07E-02	4.48E-01	1.46E-04		1.46E-04	Sn-126	1.35E+00	2.66E-01	1.61E+00
Ni-59	2.98E-01	2.73E-01	5.71E-01	7.17E-01	1.19E-02	7.29E-01	4.87E-01	1.19E-02	4.99E-01	1.63E-04		1.63E-04	Ni-59	1.50E+00	2.97E-01	1.80E+00
Nb-94	9.56E-01	2.41E+01	2.51E+01	2.30E+00	1.61E+00	3.91E+00	1.57E+00	1.61E+00	3.18E+00	5.25E-04		5.25E-04	Nb-94	4.83E+00	2.74E+01	3.22E+01
U-233	4.69E-05	4.30E-05	8.99E-05	1.13E-04	1.88E-06	1.15E-04	7.67E-05	1.88E-06	7.86E-05	2.57E-08		2.57E-08	U-233	2.37E-04	4.68E-05	2.83E-04
U-235	2.09E-02	1.92E-02	4.02E-02	1.21E-01	8.41E-04	1.22E-01	7.00E-02	8.41E-04	7.08E-02	1.15E-05		1.15E-05	U-235	2.12E-01	2.09E-02	2.33E-01
U-238	2.16E-02	1.98E-02	4.14E-02	5.20E-02	8.66E-04	5.28E-02	3.77E-02	8.66E-04	3.86E-02	1.18E-05		1.18E-05	U-238	1.11E-01	2.15E-02	1.33E-01
Totals	9.93E+04	5.23E+04	1.52E+05	2.24E+05	2.53E+03	2.26E+05	1.50E+05	2.53E+03	1.53E+05	5.45E+01		5.45E+01	Totals	4.73E+05	5.74E+04	5.31E+05

The liquid waste in WM-188 was sampled in November 2002 (sample log 021125-2) when the tank contained 214,000 gallons of waste. After the tank was sampled, it was filled with additional SBW, bringing the total waste volume to 282,000 gallons in April 2005. When the tank was sampled in November 2002, the tank contained SBW that had originally been in tanks WM-181, -184, -186, and -185 and had been concentrated in the high-level liquid waste (HLLW) Evaporator, located in the NWCF. Between 2002 and 2004, those tanks were cleaned in preparation for tank closure and the tank heels (liquids and solids) were flushed into WM-187. The liquid waste in WM-187 was then concentrated in the HLLW Evaporator, and the concentrate was sent to WM-188. This increased the WM-188 volume to 264,000 gallons by July 2004. The 50,000 gallons of waste added after WM-188 had been sampled came from the same tanks and was concentrated in the same fashion as the 214,000 gallons that were sampled in November 2002. The composition of the new waste should have been similar to that of the older, sampled waste. Thus the November 2002 waste sample is representative of the first 264,000 gallons of waste. In July 2004, concentration of the WM-180 waste in the HLLW Evaporator began. Initially, the WM-180 concentrate went into WM-188, bringing the waste volume to its current value of 282,000 gallons. Historical samples of the SBW in WM-180 (Swenson 2004b) show its composition was similar to the SBW in WM-181, -184, and -186 that had been concentrated and sent to WM-188 earlier. Because the WM-180 composition was similar to the other wastes added to WM-188, and because the WM-180 concentrate is only about 6% (18,000 gallons out of 282,000 gallons) of the total waste in WM-188, the November 2002 SBW sample should be representative of the waste currently stored in WM-188.

The activities of the gamma emitting radionuclides in WM-188 in this report differ from those of Demmer and Johnson (2003), which documents the November 2002 sampling of the WM-188 waste. In preparing this report, a review of the analytical data found the activities of the gamma emitters were suspiciously low. A review of the sample data by laboratory personnel found an error in the reported data. Correction of the error increased the activity of the gamma emitters by an order of magnitude.

The waste in WM-189 was sampled in March 2002 (sample log numbers 020313-1 and 020314-1 for radiochemical analyses) when the tank contained 282,000 gallons of waste. Since the tank was sampled, it has received an additional 2,700 gallons of waste in 2004. During that time, instrumentation calibrations reduced the measured waste volume by 2,700 gallons for a net change of 0 gallons, and the waste volume in April 2005 was 282,000 gallons. The 2,700 gallons of new waste was concentrated SBW from the HLLW and PEW Evaporators from sources similar to the SBW already in the tank. Because the new waste was similar to the existing waste and was only 1% of the total waste volume, the March 2002 liquid waste sample should be representative of the waste currently in WM-189.

The Tc-99 activity in the WM-189 liquid waste sample from March 2002 appears to be an order of magnitude too high. WM-188 and WM-189 received wastes from the same sources and thus have similar waste compositions, as confirmed by sample data. The Tc-99 activity measured in the WM-189 sample was an order of magnitude higher than in WM-188, and an order of magnitude higher than the ORIGEN2-based model estimate. A dilution error or some other sample analysis problem may have occurred that affected the WM-189 Tc-99 analysis. Based on historical SBW sample data, the WM-188 Tc-99 activity, and the ORIGEN2-based estimate, the Tc-99 activity from the WM-189 sample was not used in the Tank Farm inventory estimate.

The liquid waste in WM-190 was sampled in August 1980 (sample log number 800812-12) when several thousand gallons of contaminated water were removed from the tank resulting in its current volume of about 500 gallons. Since that time, there have been no deliberate additions of waste to the tank. There has also been no measurable increase in the waste volume in WM-190 that would indicate waste leakage into the tank. Therefore, it is presumed the composition of the waste has not changed significantly from that of the 1980 sample (other than radioactive decay).

The samples used as the basis of the Tank Farm liquid waste composition for WM-187 through WM-189 came from waste that was transferred from the Tank Farm to the NWCF, where the samples were obtained. A small amount of waste dilution occurred during the waste transfers due to the condensation of steam when transfers were made with steam jets. Therefore the liquid waste sample data were adjusted to correct for the steam jet dilution, and thus more closely reflect the waste composition in the Tank Farm. The WM-188 sample results were increased by 4.6% based upon reported sample dilution values (Demmer and Johnson 2003). The WM-187 sample had no reported dilution value, so the WM-188 jet dilution value was used, due to the similarity in system configuration between WM-187 and WM-188. The results of one of the WM-189 samples (020314-1) were increased by 2.8% based upon reported sample dilution values (Batcheller and Taylor 2003). The results of the other WM-189 sample (020313-1) were not adjusted because that waste was transferred to the NWCF via an air lift and was not diluted.

3.5.2 Radionuclides in the Tank Farm Solids

The radioactivity in the Tank Farm solids in Figure 1 is in the three 300,000-gallon waste tanks (WM-187 through WM-189) that currently store concentrated SBW. Table 2 contains data for the radionuclides included in Figure 1 and additional radionuclides of interest for each of the three tanks. Because WM-190 has never been used to store concentrated waste, it is assumed it contains no solids.

3.5.2.1 Solids Mass Estimates

Historically, INTEC controlled the composition of the Tank Farm waste to avoid the formation of solids in the tanks. Despite this, a small layer (a few inches) of solids accumulated on the bottom of each tank. The solids came from a variety of sources including insoluble material from the fuel dissolution process and some precipitation, especially from chemical additives such as the calcium nitrate used in the waste calcination facilities. The solids were light and flocculent and formed a slurry (mixture of solid particles and interstitial liquid) on the bottoms of the tanks.

There is currently no way to routinely measure the mass of solids in the tanks. Historically, the most accurate estimates of the mass of solids were made when a tank was emptied and a visual inspection of the tank interior (via camera) was performed. The 300,000-gallon tanks are 50-ft diameter right cylinders (with a volume of 1224 gallons per inch). The solids slurry is about 25 volume percent solid and 75 volume percent liquid, based on historical slurry sample data (Poloski 2000; Poloski and Wilcox 2000). The solids particle density is 2 g/cc. These data yield an equivalent volume of 306 gallons of liquid-free solids per inch of slurry, or a solids mass of 2320 kg per inch of slurry. A limited number of historical tank inspections estimated the depth of the solids slurry in some of the tanks, and consequently the mass of solids in the tanks. The observed slurry depth varied from less than an inch to as much as eight inches. However, due to the limited number of tank inspections and the varying quantities of solids in the tanks, it has been difficult to quantify the amount of solids in the entire Tank Farm.

The EIS basis report (Millet and Staiger 2000) assumed 95,100 kg of solids (equivalent to a total of 41 inches of solids slurry) were in the Tank Farm. The WIR report (Swenson 2003) assumed 85,940 kg of solids (equivalent to a total of 37 inches of solids slurry) were in the Tank Farm. The decrease in the mass of solids compared to the EIS report was based upon tank inspections made after the EIS report was generated. The SBW feed composition report (Barnes 2004) assumed 120,000 kg of solids were in the tanks, an increase of nearly 50% above that of the WIR report. The estimate in the SBW feed composition report was based upon qualitative data obtained when emptying waste tanks (such as increased probe plugging due to entrained solids when transferring waste from nearly empty tanks). The

revised estimate was not based on additional visual tank inspections. The estimate of the mass of solids is not nearly as reliable as the measured volume of liquid waste. The SBW feed report acknowledges this uncertainty by providing waste compositions that include a range of plus or minus 30% of the estimate of the solids mass.

This report assumes there are 115,000 kg of solids in the Tank Farm. This value, except for one minor adjustment, comes from the SBW feed composition report, which estimated 120,000 kg. This report assumes there are 105,000 kg in WM-187 (same as the SBW feed report after the “future” emptying of WM-180), 5000 kg in WM-188 (same as the SBW feed report), and 5000 kg in WM-189. The estimated solids mass in WM-189 differs from the SBW feed composition report estimate of 10,000 kg. Tanks WM-188 and WM-189 had similar waste storage histories and currently store similar wastes. The SBW feed report assumed WM-189 contained more solids than WM-188 because it received NWCF flush and scrub solution following the May 2000 shutdown, and because the sample from WM-189 contained more solids than the sample from WM-188.

This report decreased the WM-189 solids estimate from that of the SBW feed report based upon a review of process data and system configuration. The NWCF scrub solution sent to WM-189 after the Calcliner shutdown in May 2000 had low levels (generally less than 10 g/L) of undissolved solids (UDS). A volume of 5000 gallons of scrub solution with 5 g/L UDS would have added only 95 kg of UDS to the Tank Farm, an insignificant amount compared to the estimated 5000 kg already presumed to be there. The bulk of the NWCF post-operation flush solution (including bed dissolution solution that may have contained high concentrations of solids) went to WM-187, not to WM-189.

The waste transfer line from WM-189 is closer to the bottom of the tank than the transfer line from WM-188. The WM-189 sample may have had more solids than the WM-188 sample because the WM-189 waste transfer entrained more solids because its transfer line is closer to the bottom of the tank. Because the post-operation waste from NWCF in 2000 does not appear to have been a major source of solids to WM-189, and differences in the transfer piping out of the two tanks could account for difference in solids in the transferred waste, this report uses the same mass of solids for WM-188 and WM-189.

As part of the effort to empty, clean, and close the tanks, the liquid and solids heels in the emptied tanks (WM-180 through WM-186) were flushed into WM-187. Most of the liquid collected in WM-187 was later removed and concentrated in the HLLW Evaporator. However, the bulk of the solids flushed into WM-187 remain in the tank. As a result, the estimated mass of solids in WM-187 is much larger than that in WM-188 and WM-189.

3.5.2.2 Solids Radioactivity Estimates

The radioactivity estimates for the solids come from a limited number of solids samples. Unlike the liquid samples for which a large amount of historical data exists and there is a high degree of confidence in the accuracy of the activity estimates, it is uncertain how well the limited number of solids samples represent the composition of the solids in the tanks.

Most of the differences among the recent Tank Farm radionuclide inventory estimates were the result of varying assumptions for the mass and radionuclide activity of the solids in the tanks. The EIS basis report (Millet and Staiger 2000) assumed all solids had a Cs-137 activity of 2.4 Ci/kg (based upon a sample from WM-188 obtained in 1999). This was a conservative, bounding value and was used even though historical sample data indicated solids in other tanks had lower activity.

The WIR report (Swenson 2003) used three different radiological source terms for the solids, depending on the waste storage history of each individual tank. The WIR report used a Cs-137 activity of 2.4 Ci/kg

(the high, bounding EIS value) for only three tanks, which contained the least mass of solids. The WIR report used a lower Cs-137 activity for the solids in other tanks. It used a Cs-137 activity of only 0.43 Ci/kg for the bulk of the solids. The lower activity (along with slightly lower total solids mass) significantly reduced the Tank Farm radionuclide inventory from that of the EIS report.

The SBW feed composition report (Barnes 2004) used a different radiological source term for the solids than either the WIR or EIS reports, based upon more recent sample data. The SBW feed composition report used different values for different tanks, but it used an activity of 0.55 Ci/kg for the Cs-137 activity for the bulk of the solids (those in WM-187). The Cs-137 activity of the solids in the SBW feed composition report was higher than that of the WIR report, but still much less than that of the EIS report.

This report uses the most recent analytical data from solids samples from WM-187 and WM-188 to estimate the activity of the solids in the tanks. A solids sample from WM-187 (sample log number 040210-1) was obtained in February 2004. At the time, WM-187 contained the solids from tanks WM-182, -183, -184, -185, and -186 (plus the solids originally in WM-187). In addition, WM-181 had been emptied and an attempt was made to transfer some of the solids from WM-181 to WM-187 before the WM-187 waste was sampled. Since that time, the remainder of the solids in WM-181 along with the solids in WM-180 have been transferred to WM-187. Process data indicate there was likely not a large quantity of solids in WM-180, so its impact on the large amount of solids already in WM-187 was probably small. Therefore, WM-187 likely had the most of the solids it has today when it was sampled in February 2004. In February 2004 an effort was made to mix the solids in WM-187 by transferring solution back and forth between WM-187 and the NWCF before taking the sample. However, the success of the solids mixing effort and the degree to which the sample represents the bulk solids is uncertain.

A solid sample was obtained from WM-188 (sample log number 021125-2) in November 2002. At the time, the tank contained 214,000 gallons of SBW, about 76% of its current volume. Over the next several months, WM-188 was filled with SBW similar to the waste it already contained. Prior to storing SBW, WM-188 contained first-cycle raffinate. The bulk of the solids in the tank likely came from the first-cycle raffinate historically stored in the tank, or were formed by solution instability when the chemically different SBW was initially added to the first-cycle waste heel. The 65,000 gallons of SBW added to the tank after it contained 214,000 gallons of SBW that was sampled in November 2002 likely did not affect the solids already in the tank. The additional 65,000 gallons of new waste contained very few solids. It was also similar in chemical composition to the 214,000 gallons of waste already in the tank, so there were few if any precipitates formed by mixing the similar solutions. Because of this, the solids in the tank likely did not change significantly after the November 2002 sample was obtained. Thus the November 2002 sample likely represents the solids in the tank.

A solids sample was obtained and analyzed from WM-189 (Batcheller and Taylor 2003), but the data was not used in this report. That sample began as a slurry of solids and interstitial liquid. However, instead of filtering, centrifuging, or otherwise separating the solids from the liquids, the slurry was allowed to air dry and the resulting solids were analyzed. The air-drying process added the dissolved solids of the interstitial liquid to the original solids. The interstitial liquid contributed a significant amount of activity to the resulting solids, so the resulting solids analyses were not useful in determining the radioactivity of the original solids. Because WM-188 and WM-189 have had similar waste storage histories, it was assumed the solids in those two tanks have similar activity. Therefore the WM-188 solid sample data were applied to the solids in WM-189.

An evaluation of some of the Tank Farm solids sample data by an off-site group (Harbour et. al. 2002) assumed the air-dried-slurry sample preparation method used on the WM-189 sample was also used on other Tank Farm solids samples (specifically the samples from WM-182 and WM-183 obtained by the light-duty utility arm (LDUA) in 2000). The Harbour (2002) analysis calculated the original solids

activity by subtracting the activity contributed by the interstitial liquid in the original slurry from the air-dried solids activity. A review by INTEC laboratory personnel indicated the air-dried-slurry sample preparation method was applied to only one portion of the WM-183 sample that was used to determine the relative amounts of solids and interstitial liquid in the slurry. Air drying the slurry was not the normal process used for solids sample preparation and was not used for the chemical or radiochemical analyses of the WM-182 and WM-183 solids. Instead, the WM-182 and WM-183 solids were separated from the interstitial liquid prior to analysis. Therefore, the solids composition estimates by Harbour (2002), in which the interstitial liquid components are subtracted from the solids, are in error.

4 RADIONUCLIDES REMOVED FROM THE TANK FARM

Table 3 provides data for the activity of the major, long-lived, radionuclides that were added to the Tank Farm, remain in the Tank Farm (in both liquid and solid phases), and the percentage of the radionuclides that have been removed from the Tank Farm. The inventory in Table 3 is based upon the waste volume measured in April 2005 and the estimated solids mass previously described in this report. The activity data in Table 3 are decayed to January 1, 2012. Table 3 shows the bulk of the radionuclides have been removed from the Tank Farm, with an overall removal efficiency of 98.5%. Removal and treatment of the remaining SBW should increase the overall removal efficiency to over 99.9%.

Table 3 contains information for radionuclides (H-3, Cs-134, Tc-99, etc.) that are not in Figure 1. It also lists the individual TRU components that are compiled into a single “TRU” or “key radionuclide” category in Figure 1. The total activity in Table 3 is slightly higher than that in Figure 1 because of the additional radionuclides included in Table 3 that are not in Figure 1.

4.1 Anomalies in Radionuclide Removal Percentages

Although the removal percentages for most of the individual radionuclides in Table 3 are in the upper 90 percent range, consistent with the overall removal percentage, some radionuclides have removal percentages significantly above or below the average.

The removal percentage for Cm-244 is only 75.9%, which is much lower than other radionuclides and Cm isotopes. The removal percentage of Pu-236 (82.5%) is also well below the average. This is an artifact of using decayed data for the value of the activity generated and sent to the Tank Farm and applies to all short-lived radionuclides. Cm-244 has a half life of 18 years and Pu-236 has a half-life of only 2.85 years. The ORIGEN2-based models used to estimate the radionuclide activity were adjusted to represent fuel that was reprocessed at a specific time. The models do not accurately estimate short-lived species in wastes generated over long periods of time. The radionuclide estimate models assume an average age for each type of waste, based on SNF reprocessing history at INTEC. The waste from reprocessing Al-clad fuel is the oldest (early 1960s), followed by coprocessing, stainless steel, Zr, and finally SBW. SBW has more short-lived activity than any other waste type because it is the youngest waste and has had shortest decay time.

The Tank Farm waste inventory is entirely “new” SBW, with relatively high activities of short-lived radionuclides. The calcine, which contains the bulk of the activity that was generated and sent to the Tank Farm, consists of old waste with highly-decayed activity of short-lived species. Due to radioactive decay, the reported amount of activity generated is much lower than the actual amount. Consequently, the reported removal percentage from the Tank Farm is lower than its actual value. The Cm-244 and Pu-236 removal percentages would be higher, similar to that of other Cm and Pu isotopes, if the activity generated were not decayed.

Table 3. Summary of major radionuclides sent to the Tank Farm (generated), remaining in the Tank Farm, and the percentage removed from the Tank Farm (all activities decayed to January 1, 2012).

Radionuclide	Total Curies Generated at INTEC	Total Curies Remaining in Tanks (Liquids)	Total Curies Remaining in Tanks (Solids)	Percent of Initial Curies Removed
Am-241	9.28E+03	2.00E+02	2.49E+01	97.6%
Am-242	1.76E+01	9.51E-02	1.88E-02	99.4%
Am-243	5.96E+00	1.31E-01	2.59E-02	97.4%
Cm-242	1.51E+01	1.20E-01	1.56E-02	99.1%
Cm-243	3.62E-01	1.71E-02	3.39E-03	94.3%
Cm-244	8.31E+00	1.82E+00	1.83E-01	75.9%
Cm-245	2.04E-03	2.24E-04	4.43E-05	86.9%
Cm-246	4.48E-04	1.47E-05	2.90E-06	96.1%
Cm-247	2.47E-10	1.63E-11	3.22E-12	92.1%
Cm-248	4.15E-09	1.72E-11	3.40E-12	99.5%
Np-237	7.53E+01	1.60E+00	5.18E-01	97.2%
Pu-236	5.94E-03	8.70E-04	1.72E-04	82.5%
Pu-238	1.07E+05	1.56E+03	1.98E+03	96.7%
Pu-239	2.83E+03	1.86E+02	3.57E+02	80.8%
Pu-240	1.46E+03	8.08E+01	1.60E+01	93.4%
Pu-241	4.73E+04	1.17E+03	2.31E+02	97.0%
Pu-242	3.94E+00	5.91E-02	1.17E-02	98.2%
Pu-244	2.96E-08	3.92E-09	7.75E-10	84.1%
Cs-137	9.46E+06	1.33E+05	2.64E+04	98.3%
Ba-137m	8.95E+06	1.26E+05	2.49E+04	98.3%
Sr-90	8.42E+06	1.05E+05	1.68E+03	98.7%
Y-90	8.42E+06	1.05E+05	1.68E+03	98.7%
Tc-99	3.67E+03	5.27E+01	2.52E+01	97.9%
I-129	6.01E+00	7.84E-02	1.55E-02	98.4%
C-14	2.91E-02	3.89E-04	7.70E-05	98.4%
Cf-249	2.34E-09	9.61E-11	1.90E-11	95.1%
Cf-250	3.48E-09	4.97E-11	9.83E-12	98.3%
Cf-251	5.96E-11	1.52E-12	3.01E-13	96.9%
H-3	7.13E+03	2.55E+01	2.13E-02	99.6%
Co-60	1.67E+03	3.41E+01	1.23E+00	97.9%
Ni-63	4.36E+05	1.60E+02	3.47E+01	100.0%
Se-79	1.10E+02	1.43E+00	2.83E-01	98.4%
Sn-126	9.84E+01	1.35E+00	2.66E-01	98.4%
Ni-59	3.71E+03	1.50E+00	2.97E-01	100.0%
Nb-94	1.54E+03	4.83E+00	2.74E+01	97.9%
U-233	1.56E-02	2.37E-04	4.68E-05	98.2%
U-235	5.89E-01	2.12E-01	2.09E-02	60.5%
U-238	2.62E-01	1.11E-01	2.15E-02	49.3%
Totals (Ci)	3.59E+07	4.73E+05	5.74E+04	98.5%

Other species with removal percentages significantly below the average include long-lived U-235 and U-238 (60.5% and 49.3% removed respectively). The low uranium removal percentage is primarily the result of uranium partitioning during SNF reprocessing. The bulk of the uranium originally in the SNF was recovered in the first-cycle extraction process. Per the SNF reprocessing design, first-cycle waste contained virtually all of the fission products in SNF, but very little uranium. The second and third-cycle uranium purification processes lost about the same amount of uranium to their raffinates as the first-cycle process did. However, the second and third-cycle waste contained relatively small quantities of radionuclides. Thus, first-cycle raffinate had large amounts of radioactivity and little uranium, but second/third-cycle waste had little radioactivity and comparatively large amounts of uranium. Historically, most of the third-cycle waste and much of the second-cycle waste was stored with the SBW. The SBW remaining Tank Farm contains much of the low-radioactivity/high-uranium waste from second/third-cycle raffinate. However, the calcine contains primarily first-cycle raffinate with its high-activity/low-uranium content. This partitioning of uranium results in its relatively low removal percentage from the Tank Farm.

Although uranium partitioning is the primary reason for the reduced uranium removal efficiency, use of the SBW model to estimate uranium activity in waste for which analytical data does not exist lowers the uranium removal percentage below its actual value. A radiochemical analysis for U-235 and U-238 was performed on the WM-187 solids sample, but neither isotope was detected. Therefore, the SBW model was used to estimate the uranium activity in the solids. If one converts the estimated U-235 and U-238 activity into a mass-based concentration, the estimated uranium in the solids is about 650 mg/kg. The WM-187 solids analysis also included an elemental analysis for uranium. The elemental analysis did not detect any uranium and had a detection limit of 222 mg/kg. The elemental analysis indicates the uranium activity estimated by the SBW model is at least a factor of 3 too high. The U-234 analysis confirms the uranium activity estimate is too high. The measured U-234 activity in the WM-187 solids was 1.68×10^{-6} Ci/kg. The SBW model would estimate the U-234 activity at 7.78×10^{-6} Ci/kg, a factor nearly 5 times the measured activity. These data show the SBW model overestimates the uranium activity remaining in the Tank Farm solids, which reduces the uranium removal percentage.

A factor that affects the U-238 removal percentage, reducing it below the value for U-235, is the historical use of natural and depleted uranium (which was high in U-238 activity) in the INTEC pilot plants for SNF reprocessing studies. Wastes from those studies went to the PEW Evaporator, which concentrated the waste and sent it to the Tank Farm as SBW. Most of the U-238 from the pilot plants did not go to the first-cycle waste tanks and the calcination process. Instead, it remains in the Tank Farm as SBW. As expected, the removal percentage for U-238 is the lowest of all the radionuclides in Table 3.

The removal percentage of U-233 (98.2%) appears to be typical of most radionuclides, but it is inconsistent with the low removal percentages of the other uranium isotopes (U-235 and U-238). The U-233 originally in the SNF was removed and recovered with the other uranium isotopes during SNF reprocessing, leaving the waste depleted in U-233. However, U-233 is a daughter product of the decay of Np-237, which exists in the waste in relatively large quantities (compared to U-233). The activity of U-233 increases significantly over time (orders of magnitude over thousands of years) due to the decay of Np-237. Calcine is comprised of relatively old waste in which the decay of Np-237 has increased the activity of U-233. Due to the use of decayed activities, this increases the activity of U-233 generated and sent to the Tank Farm (even though much of it was formed in the CSSFs). The U-233 activity in the relatively young SBW remaining in the Tank Farm is comparatively low because less Np-237 has decayed to form U-233. Because the activity of U-233 in the calcine is relatively high, the resulting Tank Farm removal percentage is also high (compared to other U isotopes). This phenomena is the opposite of that previously discussed for the short-lived species (such as Pu-236) in which the activity in the calcine

has decayed more than the activity remaining in the Tank Farm, resulting in lower-than-actual reported removal percentages.

Two species on Table 3, Ni-59 and Ni-63, have unusually high removal efficiencies (100% when rounded to three significant figures). Ni-59 and Ni-63 were generated in reactors by the activation of nickel that was part of the stainless steel fuel cladding and an impurity in the Zr alloys used as fuel cladding. The activity of those radionuclides may be overestimated in the Zr-clad fuel because the Zr-clad fuel radionuclide model assumed the maximum amount of contaminants in the Zr alloy. Overestimating the activated Ni in the Zr waste model increases the activated Ni inventory in the CSSFs (because virtually all Zr waste has been calcined). This increases the total activity generated and sent to the Tank Farm, which increases the percentage removed to 100%.

The Pu-239 removal percentage in Table 3 (80.8%) is lower than other Pu isotopes and most other radionuclides. The low Pu-239 removal percentage is the result of the relatively high Pu-239 activity measured in the WM-187 solids, combined with the large amount of solids assumed to be in WM-187. Laboratory personnel performed a cursory review of the WM-187 solid sample data and found no obvious errors with the analysis. However, the WM-187 solids sample Pu-239 activity is not consistent with Pu-239 data from other Tank Farm solids samples. In February 2004, when the WM-187 solid sample was obtained, WM-187 contained the solids that had been flushed from tanks WM-182, -183, -184, -185, -186, as well as the solids originally in WM-187. It also had some, but not all of the solids from WM-181. The solids from several of those tanks had been sampled in the recent past. Table 4 shows recent solid sample results from six of the tanks. They include WM-181, -182, -183 and -186, whose solids were flushed into WM-187. It also includes a solid sample from WM-188. The solids from WM-188 were not sent to WM-187. However, due to similar waste storage histories, the solids in WM-188 were likely similar to those that existed in WM-187 before it began receiving solids from the other tanks. Table 4 also provides the February 2004 WM-187 solids sample for comparison with the other solids data. The WM-187 solids sample should have been a weighted composite of the other solids samples and been within the range of data from the other tanks.

Table 4. Comparison of solids sample data from WM-187 and other tanks whose solids were sent to WM-187.

Radionuclide	WM-181 (030209-2)*	WM-182 (LDUA)	WM-183 (LDUA)	WM-186 (010524-5)	WM-187 (040210-1)	WM-188 (LDUA)
Pu-239 (Ci/kg)	1.42E-03	1.47E-03	1.25E-03	0.73E-03	3.37E-03	0.43E-03
Am-241 (Ci/kg)	1.48E-04	8.46E-04	2.45E-04	1.09E-04	2.32E-04	2.11E-04
Np-237 (Ci/kg)	0.62E-06	1.68E-06	1.76E-06	0.86E-06	0.82E-06	2.85E-06

*Log number/source of data given beneath tank number

Table 4 shows the Am-241 and Np-237 activities in the WM-187 solids are within the activity range of the solids that were sent to WM-187. The Am-241 activity varies from 1.09 to 8.46 x 10⁻⁴ Ci/kg in the solids that were sent to WM-187. The Am-241 activity in the WM-187 solids (2.32 x 10⁻⁴ Ci/kg) lies within the expected range, and is very near the activity of the most of the tank solids samples. The Np-237 activity in the solids sent to WM-187 varied from 0.62 to 2.85 E-6 C/kg. The Np-237 activity in the WM-187 solids (0.83 x 10⁻⁶ Ci/kg) is within the range of the activity of the solids it received.

However, the Pu-239 activity (and to a lesser extent Pu-238) does not behave as expected. The Pu-239 activity in the tanks that sent solids to WM-187 varied from 0.43 to 1.47 x 10⁻³ Ci/kg. The Pu-239 activity in the February 2004 WM-187 sample was 3.37 x 10⁻³ Ci/kg, which is over a factor of 2 higher than the highest Pu-239 activity in any of the other solids, and nearly a factor of 3 higher than the average Pu-239 activity in the other tanks. This data comparison is not perfect because two tanks (WM-184 and

WM-185) that sent solids to WM-187 have no recent solid sample analysis. However, WM-184 historically held SBW similar to that of WM-181, and WM-185 held first-cycle raffinate similar to WM-182 and SBW similar to that of WM-181. Therefore, the solids in WM-184 and WM-185 were likely similar to those in WM-181 and WM-182, for which sample data exist. It is unlikely the solids in WM-184 and WM-185 were so different from those in other tanks as to change the Pu-239 activity (but not other radionuclides) from the expected value in WM-187.

Table 2 shows the Pu-239 activity in the WM-187 solids skews the Pu-239 activity distribution in the Tank Farm. The solids in WM-187 contain 12 % of the total Am-241 activity in the Tank Farm, 11% of the total Cm-242 activity, 24% of the total Np-237 activity, and 11% of the total Cs-137 activity. Most other radionuclides have similar percentages in the WM-187 solids. However, the solids in WM-187 have 65% of the total Pu-239 activity in the Tank Farm, far more than the average radionuclide. Because the Pu-239 activity in the WM-187 solids is disproportionately high compared to other radionuclides, and is higher than the activity in the solids sent to WM-187, the measured Pu-239 activity in the WM-187 solids is suspect.

Unlike historical liquid waste for which numerous samples and analyses exist that can be compared to determine anomalous sample results, there are very few solid sample analyses to compare and determine if a given sample result is erroneous. This report used the Pu-239 activity from the WM-187 solids sample, even though it appears to be high. The high Pu-239 activity in the WM-187 solids increases the Pu-239 activity in the Tank Farm, which lowers the Pu-239 removal percentage below that of most other radionuclides.

4.2 Comparison of Radionuclide Removal Data with the WIR

Table 5 compares the radionuclide data (activity generated, removed from the tanks, and remaining in the tanks) from Table 3 with similar data from the WIR report (Swenson 2003). In comparing the two sets of data, the percentage removed from Table 3 of this report is generally more consistent, with less variability among the individual radionuclides, than the data from the WIR report. The few radionuclides whose percent removal differs significantly from the average (such as Pu-236, U-235 and U-238) were discussed previously. Most of the individual radionuclides have higher removal percentages in this report than in the WIR report. This is the result of the improved estimates from the revised ORIGEN2-based source term (Wenzel 2005), fewer conservative assumptions regarding the Tank Farm waste inventory, and incorporation of the operation of the Calcliner in the year 2000 into the Tank Farm radionuclide inventory.

An example of the better percentage removal correlation among radionuclides is Np-237 whose “percent removed” went from 74% in the WIR report to 98% in this report, virtually the same as the bulk of the radionuclides. This change was due to an increase in the Np-237 activity in the calcine (and consequently activity generated), coupled with a reduction of its activity in the SBW remaining in the tanks. The calcine activity increased because the original radionuclide estimate model underestimated the activity in the calcine. The original radionuclide estimates were based upon analytical data from waste samples taken when INTEC recovered Np-237 as a product, thus generating Np-237 depleted calcine. The new inventory more accurately differentiates between Np-237 depleted and non-depleted calcine. The Np-237 in the SBW is lower because the original SBW estimate adjusted the Np-237 based on laboratory detection limits instead of actual values of Np-237 in some samples. This caused the model to overestimate the Np-237 in SBW. The improved radionuclide estimates in the new ORIGEN2-based model (Wenzel 2005) results in the Np-237 removal percentage being similar to that of other radionuclides.

Table 5. Comparison of INTEC Tank Farm radioactivity generation, removal, and current inventory (decayed to 2012) between this report and that of the WIR report (EDF-1920)*.

Radionuclide	Total Curies Generated at INTEC	EDF-1920 Total Curies Generated at INTEC	Total Curies Remaining in Tanks (Liquids)	EDF-1920 Total Curies Remaining in Tanks (Liquids)	Total Curies Remaining in Tanks (Solids)	EDF-1920 Total Curies Remaining in Tanks (Solids)	Percent of Initial Curies Removed	EDF-1920 Percent of Initial Curies Removed
Am-241	9.28E+03	9.13E+03	2.00E+02	2.90E+02	2.49E+01	7.64E+01	97.6%	96.0%
Am-242	1.76E+01	1.66E+00	9.51E-02	5.41E-02	1.88E-02	1.41E-02	99.4%	95.9%
Am-243	5.96E+00	1.28E+01	1.31E-01	7.81E-02	2.59E-02	2.04E-02	97.4%	99.2%
Cm-242	1.51E+01	1.38E+00	1.20E-01	4.50E-02	1.56E-02	1.18E-02	99.1%	95.9%
Cm-243	3.62E-01	5.00E-01	1.71E-02	7.81E-02	3.39E-03	2.04E-02	94.3%	80.3%
Cm-244	8.31E+00	2.31E+01	1.82E+00	3.90E+00	1.83E-01	1.02E+00	75.9%	78.7%
Cm-245	2.04E-03	6.63E-03	2.24E-04	1.11E-03	4.43E-05	2.91E-04	86.9%	78.9%
Cm-246	4.48E-04	4.63E-04	1.47E-05	7.21E-05	2.90E-06	1.89E-05	96.1%	80.4%
Cm-247	2.47E-10	5.49E-10	1.63E-11	8.11E-11	3.22E-12	2.12E-11	92.1%	81.4%
Cm-248	4.15E-09	6.23E-10	1.72E-11	8.71E-11	3.40E-12	2.28E-11	99.5%	82.4%
Np-237	7.53E+01	4.11E+01	1.60E+00	1.04E+01	5.18E-01	1.50E-01	97.2%	74.3%
Pu-236	5.94E-03	2.03E-03	8.70E-04	4.20E-04	1.72E-04	1.10E-04	82.5%	73.8%
Pu-238	1.07E+05	9.08E+04	1.56E+03	2.31E+03	1.98E+03	1.16E+03	96.7%	96.2%
Pu-239	2.83E+03	3.11E+03	1.86E+02	2.77E+02	3.57E+02	1.18E+02	80.8%	87.3%
Pu-240	1.46E+03	1.47E+03	8.08E+01	3.74E+01	1.60E+01	1.02E+01	93.4%	96.8%
Pu-241	4.73E+04	3.25E+04	1.17E+03	9.83E+02	2.31E+02	4.70E+02	97.0%	95.5%
Pu-242	3.94E+00	3.14E+00	5.91E-02	6.32E-02	1.17E-02	7.70E-03	98.2%	97.7%
Pu-244	2.96E-08	3.83E-08	3.92E-09	2.52E-09	7.75E-10	6.60E-10	84.1%	91.7%
Cs-137	9.46E+06	8.83E+06	1.33E+05	1.39E+05	2.64E+04	3.63E+04	98.3%	98.0%
Ba-137m	8.95E+06	8.36E+06	1.26E+05	1.31E+05	2.49E+04	3.43E+04	98.3%	98.0%
Sr-90	8.42E+06	7.77E+06	1.05E+05	1.20E+05	1.68E+03	2.53E+04	98.7%	98.1%
Y-90	8.42E+06	7.77E+06	1.05E+05	1.20E+05	1.68E+03	2.53E+04	98.7%	98.1%
Tc-99	3.67E+03	3.69E+03	5.27E+01	4.22E+01	2.52E+01	1.81E+02	97.9%	93.9%
I-129	6.01E+00	6.44E+00	7.84E-02	2.48E-01	1.55E-02	5.19E-02	98.4%	95.3%
C-14	2.91E-02	2.99E-02	3.89E-04	4.50E-04	7.70E-05	1.18E-04	98.4%	98.1%
Cf-249	2.34E-09	1.81E-09	9.61E-11	6.31E-11	1.90E-11	1.65E-11	95.1%	95.6%
Cf-250	3.48E-09	5.67E-10	4.97E-11	2.64E-11	9.83E-12	6.91E-12	98.3%	94.1%
Cf-251	5.96E-11	3.14E-11	1.52E-12	9.91E-13	3.01E-13	2.59E-13	96.9%	96.0%
H-3	7.13E+03	8.16E+03	2.55E+01	3.47E+01	2.13E-02	1.40E+00	99.6%	99.6%
Co-60	1.67E+03	1.01E+03	3.41E+01	2.86E+01	1.23E+00	4.16E+00	97.9%	96.8%
Ni-63	4.36E+05	5.10E+03	1.60E+02	1.48E+02	3.47E+01	2.75E+01	100.0%	96.6%
Se-79	1.10E+02	1.08E+02	1.43E+00	1.62E+00	2.83E-01	4.24E-01	98.4%	98.1%
Sn-126	9.84E+01	9.59E+01	1.35E+00	1.53E+00	2.66E-01	4.01E-01	98.4%	98.0%
Ni-59	3.71E+03	1.90E+02	1.50E+00	1.42E+01	2.97E-01	3.73E+00	100.0%	90.6%
Nb-94	1.54E+03	8.82E+01	4.83E+00	4.20E+00	2.74E+01	1.38E+01	97.9%	79.6%
U-233	1.56E-02	4.35E-03	2.37E-04	9.01E-04	4.68E-05	2.36E-04	98.2%	73.9%
U-235	5.89E-01	7.39E-01	2.12E-01	7.73E-02	2.09E-02	1.89E-02	60.5%	87.0%
U-238	2.62E-01	2.95E-01	1.11E-01	6.22E-02	2.15E-02	4.18E-03	49.3%	77.5%
Totals (Ci)	3.59E+07	3.29E+07	4.73E+05	5.15E+05	5.74E+04	1.23E+05	98.5%	98.1%

*Shaded values indicate an increase in the current value compared to that of the WIR report.

Table 5 shows increases were made to the removal percentages of most of the Cm isotopes, compared to the WIR report. These changes were also the result of the new radionuclide estimate models, similar to that of Np-237. The older SBW model adjusted the Cm activity based upon laboratory detection limits for Cm, which overestimated the Cm activity in SBW. The new model decreased the Cm activity in the SBW and thus increased the percentage of activity removed from the Tank Farm.

4.3 Tank Farm Radionuclide Inventory Verification

Independent verification and validation (V&V) was performed for the Tank Farm radionuclide inventory model and data used by this report. The V&V included checking all the formulas, referenced spreadsheet cells, and the modeling approach. The V&V found no significant errors, but it made a few minor

corrections and suggested some spreadsheet improvements. All findings were documented in cell comments during the V&V process. The comments were resolved with the author and changes were verified when completed.

The primary purpose of this report is to estimate the percentage of radioactivity removed from the INTEC Tank Farm's cumulative inventory for a waste determination process. Consequently only the radionuclides and not the chemical components of the waste are addressed. The following paragraphs describe the Tank Farm radionuclide inventory model and its data.

The Tank Farm inventory model consists of three Microsoft Excel workbooks that are linked together for data use. All three files must be opened concurrently in order to change the radionuclide decay date to one of twelve dates for which ORIGEN2-based radioactivity estimates exist. The three files are:

1. WIR Update R5.xls
2. Wen R5 for WIR Update R5.xls
3. Anal R5 for WIR Update R5.xls

The first file “WIR Update R5.xls” contains:

- a spreadsheet for each of the tanks WM-187, -188, -189 and -190 containing sample analysis data for liquids and solids,
- a “Current Tank Farm” spreadsheet showing a composite of the radioactivity in the four tanks that remain in service,
- spreadsheets that generate the tables and block flow diagram (Figure 1) used in this report,
- a sheet linked to the offsite shipment data,
- a sheet linked to the calcined waste inventory,
- supporting documentation sheets including the analytical sample log data and scanned pertinent portions of references: Batcheller and Taylor (2003), Demmer and Johnson (2003), and Barnes (2004).

In the spreadsheets for each of the tanks, the radionuclide inventory model uses analytical sample data where available. In general, these data are decayed to the selected decay date using first-order radionuclide decay calculations. The model makes simplifications in decaying some of the radionuclides. Five species (namely Y-90, Rh-106, Ba-137m, Pr-144, and Pr-144m) are not decayed because they are short-lived daughter products in equilibrium with a long-lived parent. Nine uranium and TRU species for which sample data often exist are also not decayed. These nine are either relatively long-lived or their activity comes from long-lived parents and does not change measurably over the relatively short remaining life of the Tank Farm. These elements are not decayed because they have a complex decay chain including both decay and buildup from the decay of other radionuclides. Estimates for all other species are determined using ORIGEN2-based estimates of reactor-produced species in SBW with normalization to the analyzed Cs-137 activity. Each of the tanks’ spreadsheets includes a column for ‘Manual Adjustments’ which overrides any other data entries. This column may be used for data input from sources other than sample data or the ORIGEN2-based estimates. The Tank Farm radionuclide inventory is constructed from analytical sample data (some species decayed, others not decayed), estimates based on ORIGEN2-based models, specific fractions of a parent isotope, or a manual over-ride value.

Recent waste sample analytical data were entered in the WIR Update R5.xls workbook by one person and the data entry was checked (100%) by a second, independent person. Some corrections of the sample data were made to those species for which the reported uncertainty exceeded the analytical result (for example a result of $10 \text{ mCi} \pm 15 \text{ mCi}$). In such cases, the data were changed to “ND” for non-detect, which the workbook ignores and substitutes an estimated activity from the SBW model.

Future addition of new sample data for the tanks is straight forward via the empty sample data columns provided for each tank. Although new data addition is straight forward, consultation with one of the authors or checkers is recommended to assure the desired (decayed) output is obtained, due to the radioactive decay simplifications previously described.

The Current Tank Farm sheet controls the selection of the decay date with a drop-down list box (cell I2) and provides links to the other two workbooks so that the dates are synchronized throughout the model. The Tank Farm radionuclide inventory workbook is designed to readily decay the Tank Farm radionuclide inventory to any one of twelve commonly-requested, specific dates. Decay to dates other than the preset twelve can not be done because ORIGEN2-based activity estimates for those nuclides for which sample data do not exist are not available for other dates.

The second file “Wen R5 for WIR Update R5.xls” contains the estimated radionuclide activities at the twelve specific decay dates and provides the linkage for these data to be used in estimating the Tank Farm inventory. This file incorporates the ORIGEN2-based radioactivity estimates of Wenzel (2005), which was previously V&V’d by Millet (2005).

The third file “Anal R5 for WIR Update R5.xls” provides the data through linked references for the radionuclide activity removed from the Tank Farm in both the offsite shipments and the calcine inventory. This file also utilizes the ORIGEN2-based source term from Wenzel (2005) to estimate unmeasured constituent activity. This file was previously V&V’d during the generation of the calcine inventory report (Staiger and Swenson 2005).

4.4 Radionuclide Inventory Model Limitations

The estimated radionuclide inventory has some inherent uncertainties. One of the uncertainties is the estimate of the mass of solids in the tank that was previously discussed. Another is the use of the ORIGEN2-based models to estimate activity for which sample data do not exist, especially in the undissolved solids. The ORIGEN2-based models were originally developed to calculate the composition of calcine formed from liquid waste. The calcining process evaporated the water in the liquid waste, leaving the dissolved constituents as the solid calcine. With only a few exceptions, all of the liquid waste components are present in the calcine in the same relative quantities as they were in the liquid waste. The exceptions include a limited number of volatile species such as H-3 and I-129 for which special allowances can be made. Therefore, the ORIGEN2-based estimates are equally valid on liquid waste or solid calcine.

The ORIGEN2-based models are not as accurate in estimating the composition of Tank Farm solids as they are the liquid waste or calcine. Use of the ORIGEN2-based models designed for liquid and calcined waste to estimate the activity of the Tank Farm solids has a comparatively large uncertainty because the undissolved Tank Farm solids are not derived directly from the Tank Farm liquid waste, nor do they have the same relative amounts of various constituents (as is the case with calcine and liquid waste).

The radionuclides in the liquid waste are present in very small quantities on a mass basis. Precipitation of solids depends on species concentration and solubility, which differ dramatically with individual elements. Solids generated by solution instability in the Tank Farm require species present in relatively high concentrations and with low solubility. The radionuclides are present in such small (mass) quantities that precipitation is unlikely for most constituents. Use of the ORIGEN2-based models to estimate radioactivity in solids assumes both the liquids and solids in the Tank Farm have the same elemental

distribution as SBW. However, some elements (and radionuclides) will be more prone to precipitation than others due to differing chemical properties. Assuming the relative activity of radionuclides in the undissolved solids formed by precipitation of SBW is the same as in the liquid waste has a large uncertainty.

It is likely that much of the activity in the Tank Farm solids came from undissolved solids from the fuel dissolution and uranium extraction process, not from precipitation of SBW. Such solids will have a different relative chemical composition and radionuclide activity than SBW. For example, Zr is a minor component of liquid SBW, yet it is a major component of the Tank Farm solids. The Zr-bearing solids came from the Zr-clad fuel dissolution and uranium extraction process, not from precipitation of the SBW. Use of a model that assumes the Zr in the solids is in the same relative concentration as in the liquid SBW will not yield an accurate Zr concentration for the solids.

The same is true for radionuclides. Historically, first-cycle raffinate had much higher U-235 activity than U-238, reflecting the reprocessing of highly enriched fuel. On the other hand, SBW has nearly the same activity for U-235 and U-238. This is due to the addition of U-238 to the SBW from the INTEC pilot plants (which used natural and depleted uranium) via the PEW Evaporator. Only a few historical samples of Tank Farm solids have isotopic uranium data. The few that exist indicate the relative uranium activity of the solids in SBW is not the same as the liquid (Swenson 1992). In general, the solids in the SBW had a much higher U-235 enrichment than the uranium in the liquid SBW. This indicates much of the uranium activity in the solids came from the fuel dissolution process, with its high U-235 activity, and not from precipitation of the SBW.

Another example where the liquid-waste-based SBW composition does not correspond to the composition of the solids is Sr-90. The activities of Sr-90 and Cs-137 were nearly equal in historical Tank Farm liquid wastes. The activities of those two isotopes were also nearly the same in historical solids samples as well. However, it appears that the process to remove solids from tanks by flushing them with water significantly altered the Sr-90 content of the solids. Samples of the liquid and solid residue taken from the cleaned tank WM-183 show the liquid residue has nearly an order of magnitude (factor of 8) more Sr-90 than Cs-137. However, the solids contain over an order of magnitude (a factor of 50) more Cs-137 than Sr-90. Assuming the sample data are correct, it appears Sr-90 leached from the solids into the residual rinse water, leaving the solids relatively depleted and the liquid relatively enriched in Sr-90. The solids sample from WM-187, which contains the bulk of the solids rinsed from the various tanks, also shows depleted Sr-90 activity in the solids. The Cs-137 activity in the February 2004 solids sample from WM-187 was 20 times higher than the Sr-90 activity. Without sample data, the SBW model would estimate the Sr-90 activity in the WM-187 solids to be nearly equal to the Cs-137 activity, an order of magnitude higher than its measured value.

At the present time, the bulk of the Tank Farm radioactivity is in the liquid. Thus, the relatively large uncertainty in the solids activity does not affect the total Tank Farm activity as much as similar uncertainty in the liquid phase would. However, if future processes are developed to separate and treat solids and liquids separately, and the solids activity becomes more important, additional sample data or studies of the solids activity should be considered to further refine the estimate of activity in the solids.

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